On the theory of resonant susceptibility of dielectric glasses in magnetic field

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The anomalous magnetic field dependence of dielectric properties of insulating glasses in the temperature interval $10mK < T < 50mK$ is considered. In this temperature range, the dielectric permittivity is defined by the resonant contribution of tunneling systems. The external magnetic field regulates nuclear spins of tunneling atoms. This regulation suppresses a nuclear quadrupole interaction of these spins with lattice and, thus, affects the dielectric response of tunneling systems. It is demonstrated that in the absence of an external magnetic field the nuclear quadrupole interaction b results in the correction to the permittivity $\delta\chi \sim |b|/T$ in the temperature range of interest. An application of a magnetic field results in a sharp increase of this correction approximately by a factor of two when the Zeeman splitting m approaches the order of $|b|$. Further increase of the magnetic field results in a relatively smooth decrease in the correction until the Zeeman splitting approaches the temperature. This smooth dependence results from tunneling accompanied by a change of the nuclear spin projection. As the magnetic field surpasses the temperature, the correction vanishes. The results obtained in this paper are compared with experiment. A new mechanism of the low temperature nuclear spin-lattice relaxation in glasses is considered.

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I. INTRODUCTION

In 1998, Strehlow et al.[1] discovered and anomalous low-temperature sensitivity of the dielectric properties of various dielectric glasses to a magnetic field. This findings continue to attract the attention both of experimentalists and theorists.[2,3,4,5,6,7,8,9,10,11] As a result of further investigations, it was found that the effect was anomalously pronounced for an insulator in the absence of magnetic impurities.

Many low-temperature properties of glasses are successfully described by a standard tunneling model.[12,13,14] In a good approximation the tunneling systems (TS) can be treated as a particle moving in a double-well potential (DWP). After observing of the anomalous glassy behavior in a magnetic field, several extensions of the standard tunneling model have been suggested.[15,16,17] In our opinion, the model proposed by A. Würger, A. Fleischmann, C. Enss[17] is the most viable. It assumes that the tunneling particle possesses a nuclear electric quadrupole moment. As a result, the particle energy acquires an extra splitting b in the crystal electric field gradient (EFG). It is important that, in general case, the local axis of EFG differ in different wells of DWP (see. Fig. 1). The magnetic field then interacts with the nuclear spin magnetic moment and results in the Zeeman splitting. This modifies nuclear spin states in each well thus affecting the tunneling system properties.

The echo experiments provide convincing evidence of the influence of the nuclear quadrupole moments on tunneling.[6,7,8,9] Recently Nagel et al.[10] investigated an isotope effect in polarization echo experiments in glasses. They observed the qualitatively different echo spectra on amorphous glycerol when hydrogen, which has a *zero* electric quadrupole moment, was substituted by deuterium, which possesses a *nonzero* quadrupole moment. Thereupon, one can conclude that the quadrupole electrical moment of the TS's is the key feature responsible for the effect.

To our knowledge, there are several theoretical studies of the nuclear quadrupole effect on glassy dielectric properties.[18,19,20,21,22] In papers[20,21] we have investigated the low temperature limit $T < b$. We have shown that in this regime the nuclear quadrupole interaction results in the breakdown of coherent tunneling due to the formation of a nuclear spin polaron state of tunneling particle. It was demonstrated that for $T < 10mK$ the saturation in the dielectric constant temperature dependence observed in a variety of glasses can be explained by that breakdown.

The case of high temperatures $T \gg b$ has been examined in Refs.[18,19,22]. Paper[19] is concerned with the many - body relaxation of tunneling system experiencing quadrupole and Zeeman splittings. Ref.[22] addresses a nonlinear behavior of dielectric permittivity at high temperatures. Paper[18] investigates the correction to the real part of the permittivity $\delta\chi$ due to the nuclear quadrupole interaction and the magnetic field. The authors of paper[18] have supposed that this correction is due to tunneling systems, which have energy splitting of the order of temperature. As a result, a very weak contribution $\delta\chi$ has been found to possess a strong temperature dependence $\delta\chi \sim 1/T^6$, which differs significantly from the experimental behavior of $\delta\chi \sim 1/T$[18,22]

In the present paper we argue that in the case $|b| \ll T$, only tunneling systems are important, which have energy splitting of the order of quadrupole splitting $\Delta, \Delta_0 \approx b \ll T$. It is shown that for vanishing magnetic field the correction to the standard dielectric permittivity

χ can be expressed as $\delta\chi/\chi \sim |b|/T$. After application of magnetic field, $\delta\chi$ first increases sharply. When the Zeeman splitting passes the quadrupole splitting, $\delta\chi$ reaches the maximum followed by a relatively smooth decrease, yet, the dependence $\delta\chi \sim |b|/T$ remains. The suggested theory agrees qualitatively with the experimental data.[3,8]

The paper is organized as follows. In Section II we introduce the tunneling model with the quadrupole and Zeeman interactions. As an example, we derive the tunneling Hamiltonian for the nuclear spin $I = 1$. Then, in Sec. III we investigate the correction to the resonant permittivity induced by the quadrupole and the Zeeman interactions. In the following sections we present numerical analysis for the resonant permittivity in the case of nuclear spin $I = 1$. For the cases $I = 1$ and $I = 3/2$, we propose an exact analytical solution for the permittivity $\delta\chi$ induced only by the quadrupole interaction. We give the qualitative explanation for the temperature and the magnetic field dependencies. In conclusion, we consider the relation of the results obtained to the experimental data. Also, a new mechanism of the low temperature nuclear spin-lattice relaxation in glasses is considered, which is due to tunneling system quadrupole interaction.

II. GENERALIZATION OF THE STANDARD TUNNELING MODEL

According to the standard tunneling model[12,13,14] at low-temperatures, amorphous solids are represented by an ensemble of tunneling systems. One can conceive a tunneling system as a particle moving in a double-well potential (DWP) characterized by the asymmetry energy Δ and the tunneling splitting Δ_0. Motion of such a particle is described by the standard two-level *pseudospin* 1/2 Hamiltonian

$$h = -\frac{\Delta}{2}\sigma^z - \frac{\Delta_0}{2}\sigma^x. \tag{1}$$

Following Refs.,[12,13,14] we assume that parameters Δ, Δ_0 obey the universal distribution

$$P(\Delta, \Delta_0) = \frac{P}{\Delta_0} \tag{2}$$

where P is a constant.

The tunneling particle possesses its own internal degrees of freedom associated with its nuclear spin $\mathbf{I}$. The tunneling particle gains Zeeman energy in the magnetic field $\mathbf{B}$

$$E_{int} = -g\beta\mathbf{B}\hat{\mathbf{I}}, \tag{3}$$

where g is the Landé factor and β is the nuclear magneton. Typically the product $g\beta B$ reaches the value $1mK$ at $B \approx 5T$.

Consider the case of the spin $I \geq 1$. In this case the nucleus can possess an electric *quadrupole* moment. It interacts with the crystal field characterized by the tensor

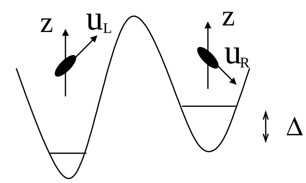

FIG. 1: Tunneling system with asymmetry energy Δ, and the corresponding quadrupole quantization axes in the left well u_L and in the right well u_R (see Ref.[17])

of electric field gradient (EFG) q_{ij}. The Hamiltonian of the particle interacting with the crystal field can be expressed as[23]

$$H_Q = b\left(I_1^2 + \frac{\varkappa}{3}\left(I_2^2 - I_3^2\right) - \mathbf{I}^2/3\right). \tag{4}$$

Here, the parameter $b = \frac{3e^{\ 2}\ Qq_{11}}{4I(2I-1)}$ designates the quadrupole interaction constant and the asymmetry parameter is given by

$$\varkappa = \frac{q_{22} - q_{33}}{q_{11}}. \tag{5}$$

We assume that the Cartesian e_1, e_2, e_3 axes are chosen so that $q_{33} \leq q_{22} \leq q_{11}$, since then $0 \leq \varkappa \leq 1$. If $\varkappa = 0$, then the EFG possesses axial symmetry. In this case, the quadrupole energy is completely defined by the spin projection I_1 and the quadrupole quantization axis is directed along e_1.

For the concreteness we will particularize the case $I = 1$ and the case of axial EFG symmetry $\varkappa = 0$. These assumptions simplify the problem noticeably. At the same time, it provides a good qualitative picture of the phenomena. In addition, some results will be presented for the case $I = 3/2$.

Let us choose the Cartesian reference system $\mathbf{e}_x, \mathbf{e}_y, \mathbf{e}_z$. Consider the eigenfunctions of the operator $I_z, |-1\rangle, |0\rangle, |1\rangle$, as a basis. Then, the Zeeman interaction, which is the same in both wells, reads

$$H_m = -g\beta B \begin{pmatrix} \cos\theta & \frac{\sin\theta}{\sqrt{2}}e^{-i\varphi} & 0 \\ \frac{\sin\theta}{\sqrt{2}}e^{i\varphi} & 0 & \frac{\sin\theta}{\sqrt{2}}e^{-i\varphi} \\ 0 & \frac{\sin\theta}{\sqrt{2}}e^{i\varphi} & -\cos\theta \end{pmatrix}. \tag{6}$$

Here the angles φ and θ assign the magnetic field direction.

Let $\mathbf{e}$ and $\mathbf{e}'$ be the axis of the *EFG* in the left and the right well respectively and α the angle between $\mathbf{e}$ and $\mathbf{e}'$. Without loss of generality one can assume that $\mathbf{e}$ and $\mathbf{e}'$ belong to the $\mathbf{x}-\mathbf{y}$ plane and $\mathbf{e}_x = \mathbf{e}$. Then, in the right well

$$H_R = H_Q(\alpha) = \frac{b}{3}\begin{pmatrix} -\frac{1}{2} & 0 & \frac{3}{2}e^{-2i\alpha} \\ 0 & 1 & 0 \\ \frac{3}{2}e^{2i\alpha} & 0 & -\frac{1}{2} \end{pmatrix}, \tag{7}$$

while in the left well

$$H_L = H_Q\left(0\right). \tag{8}$$

Thus, the Hamiltonian of the tunneling particle in the presence of the quadrupole and Zeeman splittings reads

$$H = \frac{1}{2}\cdot\begin{pmatrix} H_L & \Delta_0\cdot\mathbf{I} \\ \Delta_0\cdot\mathbf{I} & H_R \end{pmatrix}, \tag{9}$$

where $\mathbf{I}$ is a unit matrix of rank 3 and

$$\begin{aligned} H_L &= -\Delta\cdot\mathbf{I}{+}2\cdot\left(H_{LQ}+H_m\right) \\ H_R &= \Delta{\cdot}\mathbf{I}{+}2\cdot\left(H_{RQ}+H_m\cdot\right) \end{aligned} \tag{10}$$

Parameter b is supposed to be approximately independent on the position of the tunneling particle within a wells. Also, we assume this parameter is almost the same in different wells. This is justified by the experimental data for nuclear quadrupole resonance in amorphous As_2S_3 and As_2Se_3 where only about 10% line widening has been found[24]

Almost nothing is known about the angle α between the quadrupole axes in the DWP except for the case of amorphous glycerol where this angle has been found to have a specific value[10]. The analytical approach below (see. Eq. (31)) reveals that there is an insignificant dependence of the result on this angle, perhaps excluding the cases where $\alpha = 0, \pi/2, \pi$. For this reason, we assume the angle distribution to be uniform on the interval $0 \le \alpha \le 2\pi$.

In this paper, for the sake of simplicity, we are restricting our consideration to the single tunneling particle possessing a nuclear spin $I = 1$. This assumption simplifies the analysis. In general, higher spins and multiple particles are possibly involved in a single tunneling system.[19,21,25]. We assert, however, that our consideration captures the important physics. In particular, our assertion is based on the fact that we have not revealed any qualitative difference between spins $I = 1$ and $I = 3/2$. The effect of a multiparticle TLS structure seems to be insignificant at high temperature where the effect of interest is a small perturbation and we can treat the nuclear quadrupole interaction for each particle belonging to a tunneling system independently. Then, the resulting effect is reduced to a sum of single particle contributions. To take into account the complex TLS structure one should multiply the quadrupole constant b by a factor of the number of particles involved in the tunneling system.

III. RESONANT PERMITTIVITY OF A MULTI - LEVEL TUNNELING SYSTEM

In the model considered in this paper the application of the external electric field makes the asymmetry parameter field dependent

$$\Delta\left(\mathbf{F}\right) = \Delta - \mathbf{F}\widehat{\boldsymbol{\mu}} \tag{11}$$

Here $\mathbf{F}$ is the strength of the field, and

$$\widehat{\boldsymbol{\mu}} = \frac{\boldsymbol{\mu}}{2}\begin{pmatrix} -\mathbf{I} & 0 \\ 0 & \mathbf{I} \end{pmatrix} \tag{12}$$

is the dipole-moment operator of the system described by the Hamiltonian (9), so that the dipole moment of the particle in the left well is $-\boldsymbol{\mu}/2$, while in the right well it is $\boldsymbol{\mu}/2$.

In our recent paper[21] we have derived a general expression for the dielectric permittivity for the ensemble of quantum system each having a discrete energy spectrum E_m

$$\chi_{ab} = \sum_{m=1}^{2n}\left(-\mu_a\mu_b P_m\frac{\partial^2 E_m}{\partial\Delta^2} + \mu_a\frac{\partial P_m}{\partial F_b}\frac{\partial E_m}{\partial\Delta}\right). \tag{13}$$

Here E_m are the eigenvalues of the Hamiltonian (9); μ_a and F_b- are the Cartesian coordinate of the vectors $\boldsymbol{\mu}$ and $\mathbf{F}$ respectively; $n = 2I+1$, e.g., $n = 3$ for the case of nuclear spin $I = 1$; P_m is the population factor for the state E_m.

Generally, in the low frequency external electrical field $\mathbf{F} \sim \exp\left(-2\pi i\nu t\right)$ the population factor P_m can change. This process, however, is important only when the tunneling system experiences the relaxation for time τ smaller then the period of electrical field oscillations ν^{-1}. In the regime of interest, $T < 50mK$ and $\nu > 100Hz$,the time τ is large enough in comparison with the field oscillation period ν^{-1}.[26] Accordingly, the relaxation rate of TLS $\tau^{-1} \sim \nu$ is so small that the population of energy levels cannot follow the rapidly changing field $\mathbf{F}$. For this reason, the population factor remains approximately field independent.[13] Therefore, the contribution of the second term to permittivity in Eq.(13) (usually called a relaxational contribution) vanishes. So the permittivity is only due to the first term in Eq.(13) called a resonant contribution. Then the population factor P_m is given by the *unperturbed* (in the absence of the weak field $\mathbf{F}$) equilibrium distribution for the by the field TLS's

$$P_m = \frac{exp\left(-\frac{E_m}{T}\right)}{\sum_{\gamma=1}^{Z} exp\left(-\frac{E_\gamma}{T}\right)} \tag{14}$$

The contribution of a single TLS should be summed over all TLS's belonging to the system. This is equivalent to averaging the susceptibility χ_{ab} in Eq. (13) over energies, tunneling amplitudes, dipole moments, and angles between quadrupole axes and the external magnetic field (angles α, θ, φ in Eqs. (6), (7)).

Taking the average over directions and absolute values of TLS dipole moments is straightforward and we can rewrite the resonant TLS contribution in the form

$$\chi_{a,b}^{res} = \delta_{ab}\frac{\mu^2}{3}\chi, \quad \chi = \left\langle\sum_{m=1}^{2n} P_m\chi_m\right\rangle, \tag{15}$$

$$\chi_m = -\frac{\partial^2 E_m}{\partial\Delta^2}. \tag{16}$$

The parameter χ_m is a function of tunneling parameters Δ, Δ_0, the external magnetic field $\mathbf{B}$, and nuclear quadrupole interaction b in both wells. The angle bracket in Eq. (15) mean the Gibbs averaging, the averaging over distribution (2), over directions of the magnetic field φ and θ and of electrical field gradients α. Thus, the permittivity χ remains the function of temperature T, the quadrupole interaction b and the magnetic field B. Note that one can ignore the external field $\mathbf{F}$ effect on the local electric field gradient interacting with the nuclear quadrupole because the field $\mathbf{F}$ is 5 to 6 orders of magnitude smaller then the crystal field.

IV. PERMITTIVITY OF TUNNELING SYSTEMS WITH QUADRUPOLE INTERACTION. THE NUMERICAL ANALYSIS.

Below we analyze the case of high temperature, i.e., the case when

$$b \ll T. \tag{17}$$

It is known that the main contribution to permittivity of tunneling systems in glasses is due to tunneling systems having $\Delta \sim \Delta_0 \geq T$.[13,19] On the other hand, it is clearly explained in[19] that merely the magnetic field (in the absence of quadrupole interaction) does not change the permittivity. Changes can occur only if the quadrupole interaction exists. Below we argue that, in the absence of the magnetic field, the correction $\delta\chi$ to permittivity caused by the nuclear quadrupole interaction is associated with tunneling systems having $\Delta \leq \Delta_0 \leq b$.

The net quadrupole effect can be described by the difference of two permittivities $\delta\chi$, one given by Eq. (15) and the other defined by the same Eq. (15) taken at the value $b = 0$

$$\delta\chi = \chi - \chi(b = 0) \tag{18}$$

In fact, it is impossible to *"turn off"* the quadrupole interaction to experimentally investigate how the quadrupole interaction influences the permittivity, if any. However, a very strong magnetic filed should inhibit the quadrupole effect[19,20,21] This is due to the fact that the strong magnetic field creates identical local eigen states, characterized by a certain nuclear spin projection onto the magnetic field in the wells. Thus, the influence of the quadrupole effect on the permittivity can be verified experimentally by measuring the difference

$$\delta\chi = \chi_{\exp}(m = 0) - \chi(m = \infty), \quad m = g\mu B, \tag{19}$$

where m is the Zeeman splitting of nuclear spin energy levels by the external magnetic field.

The permittivity of the tunneling system is completely defined by the energy spectrum of the Hamiltonian (9). This spectrum strongly depends on the relation between the nuclear quadrupole interaction b and the Zeeman splitting m.

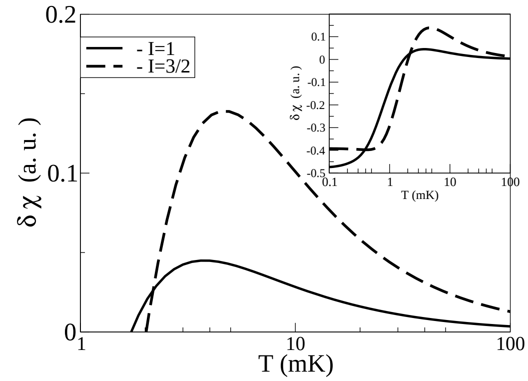

FIG. 2: A temperature dependence of the contribution to the permittivity of tunneling systems due the quadrupole interaction. The graphs clearly shows the $1/T$ dependence above the temperature $T = 5mK$. The graphs in the inset also shows a negative contribution below $T = 1mK$.

Consider first the case $m \ll b$ where one can neglect the magnetic field, i.e., approximately assuming $m = 0$. Let us first present the results of the numerical simulation for the permittivity (see. Fig. 2, 3) in the case of $b = 1mK$.These results were obtained as follows. First, we chosen a certain set of admissible values of the parameters deterring the Hamiltonian (9). Next, we found numerically the eigen values of the Hamiltonian and used Eqs. (14), (15), (16) to calculate the permittivity of a TLS with fixed parameters.[27] Then, numerical integration of the single TLS responses over all relevant parameters was performed to evaluate the effect of averaging. Fig. 2 represents the permittivity induced by the quadrupole interaction. For low temperatures $T < 5\ mK$ this correction is *negative*. Also, in this temperature range, as the temperature increased the permittivity also increased. These "low-temperature results" are due to the low temperature breakdown of coherent tunneling described in our previous papers.[20,21]

Above $5mK$ the quadrupole interaction induced permittivity falls with the temperature. The analysis reveals that this decreasing follows the dependence $|b|/T$. Figure 2 displays this dependence neatly for both spins $I = 1$ and $I = 3/2$.

Using a similar numerical approach, one can investigate the magnetic filed dependence of the permittivity. In this case, the Zeeman splitting has been taken into account in Eq. (9) with averaging the result over the direction of the magnetic field. Fig. 3 displays a sharp increase for the Zeeman splitting region $m < 2b$, then a quasi-plateau in the magnetic dependence takes place for $2b < m < T$ followed by the decrease for $2\ m > T$.

Let us now give the physical interpretation of these temperature dependencies.

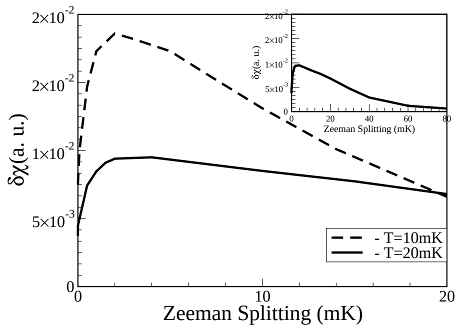

FIG. 3: Quasi-plateau magnetic field dependence of the resonant permittivity for the Zeeman splitting $5mK < m < 20mK$. The graph on the inset displays quasi exponential $\exp(-b/m)$ dependence for the Zeeman splitting $m > 20mK$.

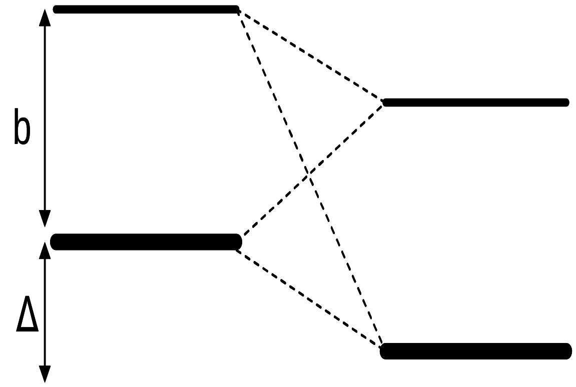

FIG. 4: The energy level structure for nuclear spin $I = 1$ in the vanishing magnetic field if the EFG possesses axial symmetry. The thick solid lines denote the degenerate levels in the left and right wells. The thin solid lines denote the non-degenerated ones. The dotted lines connect the levels coupled by the tunneling abplitude Δ_0.

A. Analytical solution for the permittivity in the case $I = 1$

There are four physically different cases $m \ll b \ll T$, $m \leq b \ll T$, $b \ll m \ll T$, $b \ll T < m$.

First, let us consider the particular case, when the magnetic field vanishes. It was assumed and then confirmed by the results obtained in this paper that only tunneling systems for which $\Delta, \Delta_0 \leq b$ significantly contribute to the correction (19). For this case, we can assume $E_m \ll T$ in Eq. (14) and, therefore, $e^{-\frac{E_m}{T}} \approx 1 - \frac{E_m}{T}$. Since for the Hamiltonian (9)

$$\sum_0^6 E_m = Sp\left(\hat{H}\right) = 0, \tag{20}$$

one can make the $1/T$ expansion of the correction

$$\delta\chi \approx \frac{\eta_1\left(\Delta, \Delta_0, b, \alpha\right)}{T},$$
$$\eta_1\left(\Delta, \Delta_0, b, \alpha\right) = \frac{1}{6}\left\langle \sum_{m=1}^{6} E_m \frac{\partial^2 E_m}{\partial \Delta^2} \right\rangle \tag{21}$$

Here the angle bracket mean the same averaging as in Eq. (15) excluding Gibbs averaging.

For the Hamiltonian (9) one can find exact expressions for E_m (see Appendix V). Also, the exact analytical averaging procedure is described in the Appendix. The final expression for the correction to the permittivity reads

$$\delta\chi\left(b, T\right) = P\frac{|b|}{T}\frac{\pi^2}{48}\left(4 - \pi\right) \approx 0.2 \cdot P\frac{|b|}{T} \tag{22}$$

This dependence shows a nonanalitic b- dependence which requires interpretation.

Correction to the permittivity $\delta\chi\left(b, T\right)$ induced by the quadrupole interaction b is due to tunneling systems having $\Delta \approx \Delta_0 \approx b$. It is evident that $\delta\chi\left(b, T\right)$ can be estimated as $\delta\chi\left(b = 0, T\right)$ taken for the tunneling systems having $\Delta \approx \Delta_0 \approx b$.Thus the correction $\delta\chi\left(b, T\right)$ is estimated to be

$$\begin{aligned} &\delta\chi\left(b, T\right) \\ &\approx P\frac{\mu^2}{3}\int_0^{|b|}\frac{d\Delta_0}{\Delta_0}\int_{-|b|}^{|b|}\frac{\Delta_0^2 d\Delta}{\left(\Delta^2 + \Delta_0^2\right)^{3/2}} \\ &\times \tanh\left(\frac{\sqrt{\Delta^2 + \Delta_0^2}}{2T}\right) \\ &\approx \frac{P\mu^2\left|b\right|}{T}. \end{aligned} \tag{23}$$

This result correlates with the numerical analysis of Sec. IV.

B. Qualitative estimate of $\delta\chi$ for intermediate, $m \approx |b|$, and strong, $m \gg |b|$, magnetic fields.

Let us return to the case when tunneling systems are described by the Hamiltonian (9) in the zero magnetic field. If one neglects the parameter Δ_0, the energy spectrum in each of the well consists of one non-degenerated level and one double-degenerated level (see, Ref.[19] and Fig. 4.

The contribution to the permittivity originates from the left and right states for which energy detuning is small compared with Δ_0. The tunneling term mutually bounds either the non-degenerate levels or the degenerate levels. In this case there exist *three* different values of Δ that result in resonance. The magnetic field eliminates degeneracy and we obtain three pairs of mutually coupled levels and there appear *six* different values of Δ resulting in resonance. Thus, the effective number of tunneling systems contributing to the resonance increases (see. Refs.[19,22])

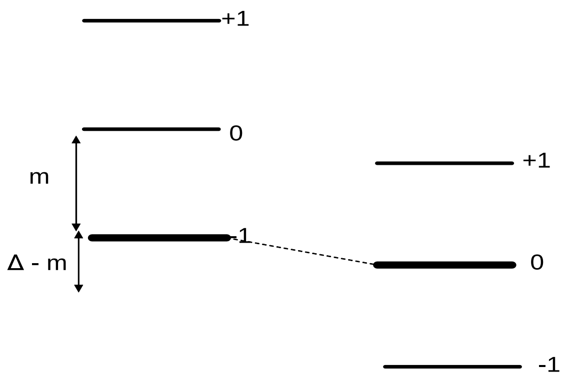

FIG. 5: The energy structure of a tunneling system in the case of a strong magnetic field $b \ll m < T$. The resonance is shown to occur between the state with nuclear spin projection $I_z = -1$ in the left well and the the state with nuclear spin projection $I_z = 0$ in the right well. The energy mismatch is $|\Delta - m| \ll m$.

In the case of a half-integer nuclear spin, energy levels in a zero magnetic field are degenerate according to the Kramers theorem. The application of a magnetic field eliminates this degeneracy. As in the case of integer nuclear spin, this results in the increase of the number of resonances. This circumstance clarifies why the application of the magnetic field increases the resonant permittivity. Note that at $T < b$ only resonance between the lowest levels in the left and the right wells is thermodynamically allowed. So the effect described above vanishes.[20,21]

Let us then turn to the case of $|b| \ll m$. In this case, one can approximately classify energy levels by nuclear spin projection I_z. If $b = 0$, the coupling between the left and right wells is possible only for levels having the same nuclear projection. If b is finite, the overlap factor between the nuclear spin states in the left and the right wells for levels with different nuclear spin projections is proportional to $|b|/m$ and can be approximated by

$$\Delta_0^{eff} \approx \frac{|b|}{m}\Delta_0 \tag{24}$$

Resonance between levels with $I_z = -1$ in the right well and $I_z = 0$ in the left well can be treated as a separate tunneling system having the energy mismatch $\Delta - m$ and the tunneling amplitude $\Delta_0^{eff} \approx \frac{|b|}{m}\Delta_0$. One can estimate its contribution to the resonant permittivity as[13,14]

$$\begin{aligned} &\delta\chi\left(m, b, T\right) \\ &= P\frac{\mu^2}{3}\int_{|b|}^{m}\frac{d\Delta_0}{\Delta_0}\int_{-\infty}^{\infty}\frac{\left(\frac{b}{m}\Delta_0\right)^2}{\left((\Delta - m) + \left(\frac{b}{m}\Delta_0\right)^2\right)^{3/2}} \\ &\times \tanh\left(\frac{\sqrt{(\Delta - m) + \left(\frac{b}{T}\Delta_0\right)^2}}{2T}\right) \\ &\approx \frac{P\mu^2}{3}\frac{\pi\,|b|}{T} \end{aligned} \tag{25}$$

This expression results in the following remarkable conclusion. In the case under consideration, the correction to permittivity is independent of the magnetic field. This conclusion does not completely correspond to the numerical simulation (see. Fig. 3) where a smooth temperature decrease in $\delta\chi$ was found. Therefore the result given by Eq. (25) needs to be amended. While writing Eq. (25) we did not take into account the population of the level with the nuclear spin projection $I_z = 0$ in the right well. This level is separated by an energy gap of the order of m from the level $I_z = -1$ in the right well which is the ground state of the tunneling system (see Fig. 5). For this reason, the population of the level $I_z = 0$ in the right well is approximately $exp\left(-m/T\right)$. Therefore, the result given by Eq. (25) should be multiplied by the factor $exp\left(-m/T\right)$ resulting in a slow decrease of $\delta\chi$ with increasing m in the region $5mK < m < 30mK$. Comparing Eq. (22) and Eq. (25) one concludes that the correction to the permittivity induced by the magnetic field is several times larger than for no magnetic field correction. This agrees with the numerical simulation presented above in Fig. 3, where the magnetic field first causes a sharp increase in the permittivity followed by a slow decrease. It is clear that the contribution to permittivity given by Eq. (25) is due to the transition between levels having different spin projections. This means that the contribution to permittivity is associated with the tunneling accompanied by a change of the nuclear spin projection. This effect is discussed below as a new potential mechanism for the spin lattice relaxation.

If the magnetic field is strong enough that $m \gg T$, only the lowest Zeeman levels are occupied what brings us and we come to the case of the standard tunneling model. In this case, the quadrupole interaction effect disappears in agreement with Fig. 5

V. CONCLUSION

It is shown in this paper that at temperatures $T > 10mK$ the correction to permittivity induced by the quadrupole splitting b behaves as $|b|/T$. This dependence correlates with the experimental data .[18] This comparison with experimental data can be used to estimate the constant $|b|$. On the other hand, one can extract from the

nuclear quadrupole resonance (NQR) experiment the frequency of the transition ω_0. If the tunneling particle carries several atoms, the effect of the number of the atoms is additive within the perturbation approach. Then, the ratio $|b|/\hbar\omega_0$ approximately estimates the number of atoms per a tunneling system.

Our results exhibit a good qualitative agreement with experiment: there is a sharp increase in permittivity as the magnetic filed is applied, followed by slow decrease after the Zeeman splitting passes the value of quadrupole splitting (see Fig. 3). In fact, in $BK7$ glass a similar effect takes place with position of the maximum at the magnetic field of the order of Teslas.[3] This agrees with the results obtained in the present paper. On the other hand, in $BaO - Al_2O_3 - SiO_2$ glass the effect is observed at magnetic field about three orders of magnitude smaller.[2,6] The reason for this effect is not clear. This can be due to the presence of residual paramagnetic impurities interacting with tunneling systems since the electronic spin in the magnetic field of the order of a few milliTeslas. acquires the Zeeman energy comparable to the temperature.

However, one can suggest an alternative interpretation of the effect in $BaO - Al_2O_3 - SiO_2$. In Ref.[19] we have shown that resonant pairs of tunneling systems experiencing quadrupole splitting noticeably contribute to dielectric loss (imaginary part of the permittivity). It has been shown that the behavior of these pairs resembles the behavior of two-level tunneling systems. The effective energy splitting for them is of the order of $10\mu K$. This value corresponds to the Zeeman splitting induced by the magnetic field of the order of milliTeslas. Thus, the magnetic field of the order of milliTeslas influences the behavior of tunneling pairs affecting the imaginary part of χ by means of change in TLS relaxation rate.[19] Because of the Kramers - Kronig relation, this circumstance should be reflected in the real part of permittivity.

According to the experiment[2,4,6], the position of the maximum in the dependence of the dielectric constant $\delta\chi$ on the magnetic field is sensitive to the value of the external electric field used in the measurements of the permittivity. This suggests the nonlinear character of the effect. The interpretation of the nonlinear effect has been proposed in Ref.[22] It was found that the maximum position is proportional to the electric field. The dependence on the electric field qualitatively resembles the behavior in Fig. 3. In the present work, we investigated only the linear response. So, to attain the linear regime, one should experiment at lower electric fields. The indication to the linear response is the peak position independence of the electric field(see Fig 3).

It is interesting that the correction to the permittivity at large magnetic fields $m > b$ is caused by tunneling accompanied by a change in the nuclear spin projection, Fig. 5. We are justified to anticipate that at low temperature a similar processes can contribute to spin - lattice relaxation with the rate exceeding that for dielectric crystals by many orders of magnitude. This new spin - lattice mechanism should be sensitive to the resonant transition rate. We expect that this transition is induced by spectral diffusion[28] which makes the transition an irreversible one. This relaxation can be slow down by the nuclear spin diffusion needed to bring a nuclear spin to the TLS neighborhood.[29] The analysis of this effect is beyond of the scope of this paper.

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APPENDIX

For the Hamiltonian (9) one can find the exact expression for E_m. The eigen energies of the Hamiltonian (9) in the case $I = 1$ at zero magnetic field read

$$E_{1,2} = b\left(\frac{1}{3} \pm \frac{E}{2}\right),\ E_{3,4,5,6} = b\left(-\frac{1}{6} \pm \frac{\sqrt{M \pm Y}}{2}\right), \tag{26}$$

$$E = \sqrt{(\Delta/b)^2 + (\Delta_0/b)^2}, M = 1 + E^2,$$
$$Y = 2\sqrt{N}, N = E^2 - (\Delta_0/b)^2 \sin^2\alpha.$$

Substituting these expressions into Eq. (21) after simplification one obtains

$$\eta_1(\Delta,\Delta_0,b,\alpha) \approx \frac{(\Delta/b)^2(\Delta_0/b)^2\left(E^2+1-4N\right)\sin^2\alpha}{6E^2N\left(4N-(E^2+1)^2\right)}. \quad (27)$$

Note that, if $\alpha = 0$ or $\alpha = \pi$, the quadrupole effect disappears in agreement with Eq.(27).

Then the total permittivity is given by the expression

$$\delta\chi(T,b) = \frac{P|b|}{T}\frac{\mu^2}{3}\frac{1}{2}\int_0^\pi d\alpha\int_0^{T/|b|}\frac{d\Delta_0}{\Delta_0}\int_{-T/|b|}^{T/|b|}d\Delta \times\eta_1(\Delta,\Delta_0,b,\alpha). \quad (28)$$

To calculate the last integral over Δ,Δ_0 it is convenient to change the variables as follows.

$$\Delta = E\cdot\sin\varepsilon,\ \Delta_0 = E\cdot\cos\varepsilon; |\varepsilon|\le\pi/2 \quad (29)$$

Then the correction (28) takes the form

$$\eta_1(E,\varepsilon,b,\alpha) = \frac{c^2\cdot\sin^2\varepsilon\cdot(M-4N)}{6g^2\cdot(4N-M^2)},\quad g=\sqrt{1-c^2},\ c=\sin\alpha\cdot\cos\varepsilon. \quad (30)$$

The indefinite integral over a parameter E reads

$$\int\eta_1(E,\varepsilon,b,\alpha)\,dE = \frac{c}{12}\sin^2\varepsilon\cdot\left(\arctan\left(\frac{2\cdot E\cdot c}{1-E^2}\right)\cdot\left(1-\left(\frac{c}{g}\right)^2\right)-\frac{2\cdot c}{g}\cdot\operatorname{arctanh}\left(\frac{2\cdot E\cdot g}{1+E^2}\right)\right),$$

The contribution of the second term in the rhs of Eq. (31) into the integral (28) vanishes since it turns to zero on the both limits $E = 0,\infty$.

To calculate the integral (28), one can substitute the integral limit $T/|b|$ by ∞ because of the fast integral convergence. The first term in the rhs of Eq. (31) is a broken function of the argument E at the point $E = 1$. Therefore, integration should be performed independently over the two intervals $(0,1)$ and $(1,\infty)$. After integrating over E one obtains the intermediate result

$$\eta_1(\varepsilon,\alpha) = \frac{\pi\cdot c}{12}\sin^2\varepsilon\cdot\left(1-\left(\frac{c}{g}\right)^2\right).$$

The indefinite integral over ε reads

$$\int\frac{\eta_1(\varepsilon,\alpha)}{\cos\varepsilon}\,d\varepsilon = \frac{\pi}{12}\left(\cot\alpha\cdot\left(\arctan\left(\frac{\tan\varepsilon}{\cos\alpha}\right)-2\arctan\left(\frac{\sin\varepsilon}{1+\cos\varepsilon}\right)\cdot\cos\alpha\right)-\sin\varepsilon\cdot\cos\varepsilon\cdot\sin\alpha\right).$$

and

$$\eta_1(\alpha) = \int_{-\frac{\pi}{2}}^{\frac{\pi}{2}}\frac{\chi_1(\varepsilon,\alpha)}{\cos\varepsilon}\,d\varepsilon = \frac{\pi^2}{12}\cdot\frac{|\cos\alpha|}{\sin\alpha}\cdot(1-|\cos\alpha|). \quad (31)$$

Finally, after the integration over the angle α one obtains

$$\delta\chi(T,b)/\chi = \frac{|b|}{T}\frac{\pi^2}{48}(4-\pi)\approx 0.2\cdot\frac{|b|}{T}.$$

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